Magnetic susceptibility: An easy approach to the spin-reorientation transition

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The reorientation of magnetization is studied during film growth by means of ac susceptibility. The susceptibility is obtained via the magneto-optic Kerr effect. For Co/Au(111) a maximum of the susceptibility is found at 4.38 ± 0.07 atomic layers of Co. The susceptibility peak is demonstrated to represent the spin-reorientation transition. The influence of external fields on the transition is explored. In agreement with theory shifts of the susceptibility peak are observed in bias fields.

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Fundamental experiments in magnetism deal with phase transitions. A well-known example is the transition from ferro-to paramagnetism which is commonly observed via the magnetic susceptibility. The susceptibility is defined as the response of magnetization due to the alteration of a magnetic field. Close to the Curie temperature T_C the magnetization tends to zero on the average as the ordering effect of the exchange interaction is compensated by the disconcerting action of thermal fluctuations of the spins. Above T_C, the ferromagnet becomes paramagnetic. This transition features a very well-pronounced susceptibility peak[1–4] that can be easily detected.

It is only recently that the susceptibility has been used to investigate the spin-reorientation transitions of ultrathin films.[5–8] Such transitions are of a somewhat special, orientational type, yet their description has been demonstrated to comply with the general framework of mean-field theory, at least.[9] The change of symmetry here results from the interplay of different anisotropy contributions whose magnitude can be controlled (driven) by temperature, thickness, or even composition variations.[10,11] Since the distinct contributions would typically vary in different ways under changes in the driving parameter, the shift in the subtle energetic balance shows up as a change in orientation of magnetization, while its magnitude remains constant as the system is still far from the corresponding T_C and deep in the ferromagnetic phase. This change in the preferred axis of magnetization direction is considered as reorientation transition.

So far only the temperature dependence of susceptibility has been studied within the context of reorientations in ultrathin films. In contrast, here we take up the opportunity to study thickness dependence of such transitions via monitoring the susceptibility during film growth by means of the magneto-optical Kerr effect. We demonstrate the power of the technique by investigating the spin-reorientation transition of Co/Au(111).

The susceptibility is measured in an alternating magnetic field (modulation field). Generally, it is a tensor. If the direction of the modulation field and the magnetization are perpendicular/parallel to each other, the transverse/parallel susceptibility will be obtained, respectively.[1,4,12] The transverse susceptibility is most sensitive to the spin reorientation as it depends on the strength of the anisotropies or, equivalently, to the stiffness of the spin alignment. Ideally, a singularity of the transverse susceptibility should be expected whenever the angle dependent energy exhibits a flat minimum as a function of the magnetization orientation, since then even very small magnetic fields will cause large swings of the magnetization direction, leading to a huge transverse susceptibility. Our experimental facilities offer the opportunity to study the magnetic ac susceptibility during thin film growth in residual fields. Hence, an in vivo study of the thickness-dependent reorientation transition can be performed.

Additionally a bias field can be applied during the susceptibility measurement. External fields affect the energetic balance of the different anisotropy contributions for the same physical reason which is operational in sensing the magnetic response, namely, the Zeeman coupling. The applied-field effect can be equivalently seen as the action, and balance, of torque, acting on the magnetization vector. This torque would typically drive the magnetization out of its zero-field orientation. Consequently, the bias field affects the spin-reorientation transition and, since the various anisotropy contributions are so very sensitive to thickness variations, a shift of the critical thickness can be expected. A field applied perpendicular to the film plane stabilizes a perpendicular magnetization direction and the reorientation transition moves into the thickness range where an in-plane orientation of the magnetization would be found without field. For Co/Au(111) this means that the spin-reorientation transition will be shifted to higher thicknesses as the magnetization reorients from perpendicular to in-plane alignment upon thickness increase.[13–17] With in-plane (horizontal) bias fields the thickness of the transition for Co/Au(111) shifts to the opposite direction of lower thicknesses. This scenario was predicted by Millev *et al.*[18] but has not been observed yet. As the magneto-optical susceptibility experiment can easily be performed in a bias field we can check this hypothesis.

Sample preparation and experiments were performed *in-situ* under ultra high vacuum (UHV) conditions ($p < 1.0 \times 10^{-10}$ mbar). The Au(111) surface was prepared by Ar ion sputtering (30° incidence) and subsequent annealing at 650 °C. The quality of the surface was checked by Auger-electron spectroscopy and low energy electron diffraction (LEED). The $23 \times \sqrt{3}$ reconstruction of the Au(111) surface[19]

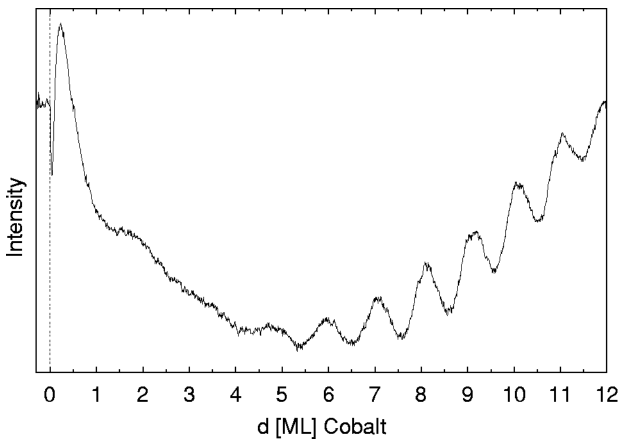

FIG. 1. MEED intensity oscillations of the specular beam during Co growth on Au(111).

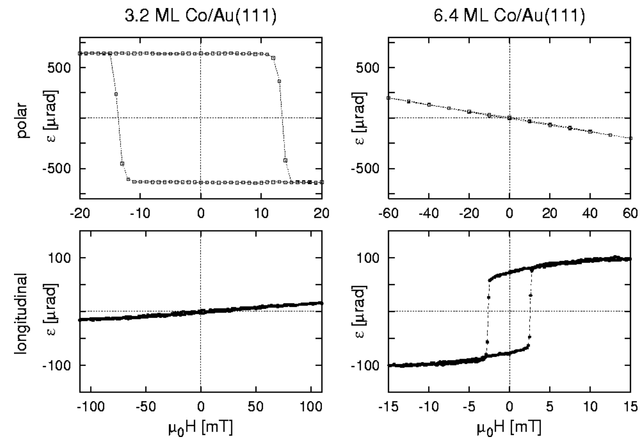

FIG. 2. Hysteresis curves at 3.2 ML (left) and 6.4 ML (right) obtained in fields perpendicular to (top) and within the film plane (bottom), i.e., polar and longitudinal MOKE geometry, respectively.

was always clearly visible in the LEED pattern. The cobalt films were grown by electron beam evaporation at room temperature at a rate of 0.3 ML/min (ML=monolayer).

The magnetic properties of the films were probed by means of the magneto-optical Kerr effect (MOKE). The experimental setup is similar to the one used by Bader *et al.*[20] The polarization change of *s*-polarized He-Ne-laser light reflected from the sample is analyzed by an arrangement of a $\lambda/4$ plate, a linear polarizer and a photodiode. *S*-polarized light is used to keep the influence of the transverse Kerr effect as low as possible.[21] The magneto-optical response is detected as a relative intensity change. It has been shown that this intensity change is proportional to the Kerr ellipticity[22] which scales linearly with the magnetization components perpendicular to the light polarization.[23,24] The $\lambda/4$ plate was incorporated in order to compensate the birefringence of the UHV windows and thus to increase the sensitivity.[25]

Magnetic hysteresis loops were obtained in magnetic fields oriented within and perpendicular to the sample plane via the longitudinal and polar Kerr effect. The angles of incidence were 45° and 15°, respectively.

The experimental setup is slightly modified for measuring the susceptibility.[5,26] Via coreless coils magnetic ac fields can be generated either within or perpendicular to the sample plane. The field strengths are some tenth of mT. The frequency is fixed to 113.94 Hz. The magnetic ac susceptibility is obtained by detecting the phase locked response of the system during Co growth via MOKE. As the frequency of the modulation field is quite low, the susceptibility can be treated as quasistatic. The susceptibility experiments are performed with bias fields applied within the film plane, parallel to the in-plane modulation field, and parallel to the plane of incidence of the light.

The growth rate was calibrated via medium energy electron diffraction (MEED). The intensity of a specular beam versus thickness is shown in Fig. 1. Modulations of the reflectivity are attributed to layer-by-layer growth[27] which are found to set in after deposition of ≈ 6 ML Co. Immediately after every susceptibility experiment the growth rate was calibrated by MEED. The Co thickness was calculated from the deposition time utilizing the average time interval between the maxima of intensity at higher thickness.

In Fig. 2 hysteresis loops for Co/Au(111) are plotted for different orientations of the magnetic field. On the left/right-hand side loops of a 3.2/6.4 ML film are shown, respectively. The magnetic field was applied perpendicular to the sample plane (polar geometry) for the upper curves and in-plane (longitudinal geometry) for the lower curves. At 3.2 ML the loop in polar geometry exhibits a rectangular shape which indicates a switching of the magnetization between the two orientations parallel to the field. The curve in longitudinal geometry exhibits hard axis behavior, i.e., a linear dependence on field. Thus the hysteresis loops demonstrate that the easy axis of magnetization is perpendicular to the film plane. At 6.4 ML the polar and longitudinal hysteresis curves have exchanged their shape with respect to the 3.2 ML case. The longitudinal hysteresis loop is rectangular whereas the polar is linear and cannot be saturated. This indicates that the preferred axis of magnetization is in-plane.

The set of hysteresis curves reveals that the magnetization is perpendicular at 3.2 ML and in-plane at 6.4 ML. In between the preferred axis switches from the perpendicular to the in-plane direction upon thickness increase, i.e., a reorientation transition occurs. Further insight is gained by the in vivo susceptibility experiments.

Figure 3 shows the susceptibility during Co growth. Only the in-phase response is displayed as the out-of-phase components are of no importance in the context of this paper. ac fields have been applied within ($H_{=}$) and perpendicular to the sample plane ($H_{\perp}$) with amplitudes of 0.12 mT and 0.05 mT, respectively. The Kerr ellipticities have been normalized with respect to the amplitudes of the modulation fields. As the polar Kerr signal is about 8 times stronger than the longitudinal Kerr signal,[28] a non-negligible contribution can be expected when probing the system with an in-plane ac field. For that reason we did not convert the signals into SI units.

The main features of the magneto-optic signals at both modulation fields are well-pronounced peaks which appear apparently at the same thickness of (4.38 ± 0.07) ML (Fig. 3). Clearly, the peaks are located within the established thickness range and can be attributed to the spin reorienta-

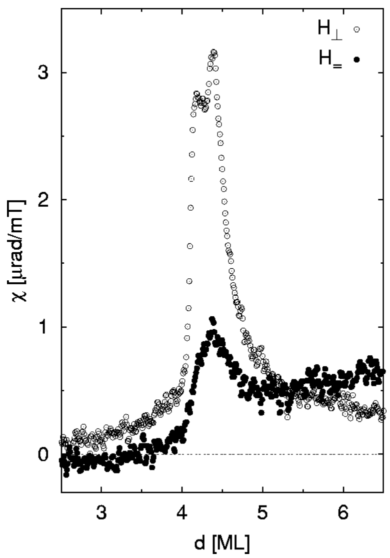

FIG. 3. The in-phase component of the magnetic susceptibility measured during growth of Co/Au(111). The ac modulation field of 0.12 mT/0.05 mT was oriented within ($H_=$) or perpendicular to the sample plane ($H_\perp$), respectively.

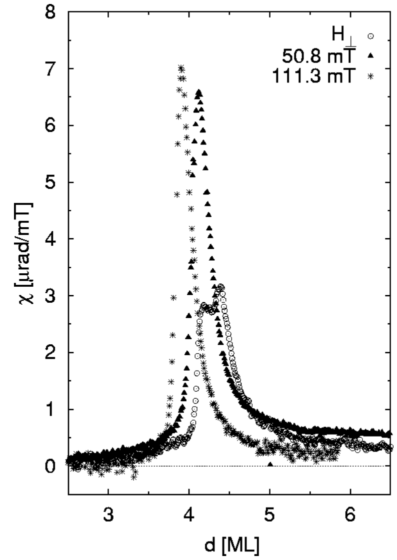

FIG. 4. The in-phase component of the susceptibility during growth in different bias fields. The values of the bias fields are given in the plot. The modulation field was perpendicular to the film plane.

tion. Different critical thicknesses for the spin-reorientation transition of Co/Au(111) have been reported in the literature as, e.g., 3.7–4.10 ML,[29] 4.3–4.7 ML,[30] and 3.5–5 ML.[31] One reason for the spread in data for the critical thickness for reorientation is the surface diffusion of Au even at room temperature. The Au coverage of the Co film increases with time[32] which increases the interface anisotropies,[33] causing shifts of the thickness of reorientation.[29,32]

Figure 3 reveals that the shape of the susceptibility depends on the orientation of the modulation field. At higher thicknesses the evolution of the two susceptibilities is different. While for perpendicular modulation the susceptibility continuously decreases, it increases with thickness in the in-plane field. The latter signal can be attributed to domain wall motion. Magnetic ac fields lead to back and forth movement of domain walls that causes the alternation of the magnetization averaged over the laser spot. Hence, a nonzero constant parallel susceptibility has to be expected. As the magneto-optical Kerr effect depends linearly on thickness in ultrathin films,[34,35] the increase of the magneto-optic response with thickness is solely a magneto-optic effect. These considerations explain the parallel susceptibility above 4.6 ML. The decreasing transverse susceptibility, on the other hand, is in agreement with the hypothesis that the effective in-plane anisotropy becomes larger on thickness increase. Consequently the upper limit of the reorientation range can be confined to 4.6 ML.

Thus we may conclude that the susceptibility peaks in the two different modulation fields are to be correlated with the spin-reorientation transition. The shapes and the detailed interpretation of the susceptibility signals will be discussed in a forthcoming paper.

The second goal of this paper is to demonstrate the influence of external fields on the reorientation transition. Theoretically, a bias field will shift the reorientation transition to other thicknesses.[18] For Co/Au(111) a horizontal bias field should shift the reorientation transition to lower thicknesses. Hence, the peaks in the ac susceptibilities should shift to lower thicknesses as well.

In Fig. 4, the magnetic susceptibility obtained in different horizontal bias fields is plotted. The modulation field (0.16 mT) was applied perpendicular to the sample plane. It is obvious that the peaks shift towards lower thicknesses in field. On field increase the shift gets larger and the peaks appear at lower thicknesses which is in complete agreement with the theoretical predictions of Millev *et al.*[18]

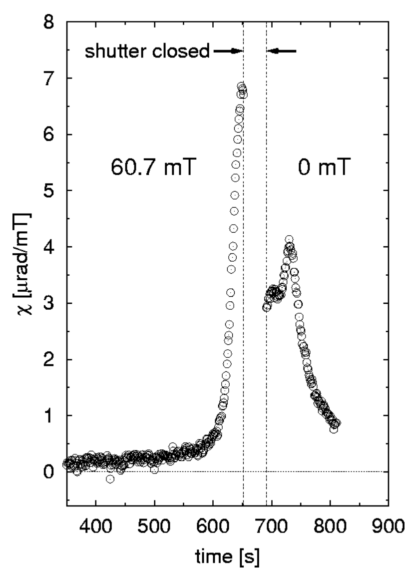

FIG. 5. In-phase component of the susceptibility during Co growth in perpendicular modulation field (0.16 mT). In the beginning a bias field of 60.6 mT was applied. After the peak the evaporation was interrupted and growth continued in zero field. For further details, see text.

To resolve the thickness shifts a combination of bias field and zero field susceptibility experiments was performed that allows to measure the relative shift of the susceptibility peaks in field (see Fig. 5). Starting with in-plane bias field, growth is interrupted immediately after the susceptibility peak and continued in zero field. The zero field peak is then observed with a certain time delay. Calibrating the growth rate by MEED after the susceptibility experiment allows then the transformation of the time delay into a thickness increase. As a result, the relative shifts of the susceptibility peaks in horizontal field with respect to zero field were determined with high accuracy. Shifts of (-0.26 ± 0.06) ML, (-0.29 ± 0.06) ML, and (-0.44 ± 0.1) ML were found for 50.8 mT, 60.8 mT, and 111.3 mT, respectively.

To compare quantitatively the field shifts more experiments particularly in bias fields oriented perpendicular to the film plane are necessary. With the additional set of data a classification of the spin-reorientation transition and a determination of anisotropy values should be possible.

The susceptibility experiments in bias fields offer the direct and easy proof of the influence of the field on spin reorientation. The interpretation of hysteresis loops at different thicknesses, particularly to the purpose of determining the critical thickness, is not straight forward, because the variation of two parameters, thickness and field, is involved. Only careful analysis of the hysteresis loop can give the correct interpretation of the switching behavior of the magnetization in field[28,36] which has to include the field effecting the spin-reorientation transition.

In summary, we have presented a simple, versatile and fundamental method to study the reorientation of the magnetization during growth. Zero-field susceptibility experiments on Co/Au(111) were performed at different modulation field orientations. The arising peaks have been shown to correlate with the spin-reorientation transition. The application of an external bias field within the film plane causes a shift in the magnetic susceptibility maxima towards lower thicknesses in conformity with the theoretical predictions.

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